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Fenitrothion and Aminocarb Residues in Water and Balsam Fir Foliage following Spruce Budworm Spraying Programs in Quebec, 1979 to 1982

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Fenitrothion (O,O-dimethyl-O-4-nitro-m-tolyl phosphorothioate) and aminocarb (4-dimethylamino-m-tolyl N-methylcarbamate) have been used operationally since 1969 and 1975 respectively for controlling the spruce budworm (Choristoneura fumiferana Clem.) in Canadian forests. Several studies had previously been carried out to determine the levels of residue left by these insecticides in water and in conifer foliage following aerial spraying in New Brunswick, Newfoundland, Ontario and Manitoba. Fenitrothion concentrations reaching 0.5 to 8.0 ug/g (dry weight) (Mallet and Cassista 1984; Yule and Duffy 1972; Yule and Varty 1975) and aminocarb concentrations of 2.7 ug/g (wet weight) (Sundaram and Szeto 1984) were reported in conifer foliage. Maximum levels of 20.0 ug/l (Mallet and Volpé 1982) and 51.6 ug/l (Holmes et al. 1984) of fenitrothion were reported for lentic water while maximum reported concentrations of the same insecticide in lotic water varied from 1.3 to 75.5 ug/l (Eidt and Sundaram 1975; Flannagan 1973; Mallet and Cassista 1984) with one study reporting levels of up to 701 ug/l (Moody et al. 1978). Maximum residue levels of aminocarb reported were 4.7 to 53.0 ug/l in lentic water and 3.3 to 24.0 ug/l in lotic water (NRCC 1983). These concentrations were detected after spraying up to 280 g AI/ha of fenitrothion and 70 or 88g AI/ha of aminocarb.

In Quebec, environmental surveillance of aerial spraying carried out from 1979 to 1982 through the analysis of insecticide residues provides a general pattern of contamination in the sprayed areas. The present paper examines the results of these analysis and we will also discuss the effect of such concentration levels on the environment.

MATERIAL AND METHODS

From 1979 to 1982, annual spraying operations covered areas of 5.8, 1.9, 7.1 and 13.0 (x 10^5) hectares, of which more than 85% were treated with chemical insecticides sprayed by four-engine aircraft. The types of aircraft used were Constellation L-749, DC-4G and DC-6, guided by a Litton inertial navigation system, and equipped with 154

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flat-jet nozzles. Sumithion™ (fenitrothion) at 210g AI/ha and Matacil 180D™ (aminocarb) at 52g AI/ha were sprayed in volumes of 1.12 liter/hectare in 1979 and 1.4 liter/hectare from 1980 to 1982. Diesel oil (insecticide diluent 585) was used to dilute both insecticides.

Water samples and balsam fir (Abies balsamea (L.) Mill.) foliage samples were collected in open areas shortly after the insecticide cloud had settled and used to detect the highest concentrations of residue. In each of the blocks sprayed, one water sample was taken from a small lake and one from a stream directly exposed to several spray lines. Samples were collected in 1-liter glass bottles from the water's surface one hour after spraying. Samples were then stored on ice in the dark until the insecticide was extracted in the laboratory. In each block sprayed, one balsam fir foliage sample was taken along the edge of a forest road under a spray line. This sample was composed of some 30 branch tips approximately 30 cm long, collected from 10 to 15 trees one to four hours after spraying. Branches were cut off one and two meters above the ground and placed in a bucket lined with aluminum foil. Needles were then taken from the current year shoots as well as those of the two previous years, mixed and transferred into a glass jar. Samples were kept frozen pending analysis.

Aminocarb present in water and foliage was analyzed according to the method described by Mamarbachi (1980). The same method was used for fenitrothion found in water, except that the pyrex column used in the gas chromatograph was 180 cm long and the temperatures of the oven and injector were 190°C and 250°C respectively. As for the analysis of fenitrothon in foliage, extraction and clean-up were conducted according to the methods described by Yule and Duffy (1972), determination by gas chromatography being the same as for water analysis.

RESULTS AND DISCUSSION

Concentrations of fenitrothion and aminocarb in water and foliage show very high variations between samples, with low concentrations in most cases and very high residue concentrations in some (Table 1). Due to such variations, median values are used rather than average arithmetic values since the former are more representative of the concentrations generally expected in the environment after spraying. Over the four years studied, median residue levels found were 3.81 ug/g (dry weight) of fenitrothion and 0.94 ug/g of aminocarb in balsam fir foliage, with maximum concentrations of 111 and 37.3 ug/g respectively. In lentic water, median residue levels were 5.82 ug/l of fenitrothion and 0.74 ug/l of aminocarb, with maximum levels of 1,114 and 331 ug/l. In lotic water, median concentrations were 2.84 ug/l of fenitrothion and 0.86 ug/l of aminocarb, with maximum levels of 127 and 18.4 ug/l respectively.

Aminocarb residue levels were always lower than those of fenitrothion since the latter is sprayed at dosage rates four times greater. Median residue concentrations for a given year were not more than 60%

TABLE 1: Fenitrothion and Aminocarb residues detected in water samples and balsam fir foliage samples collected inside blocks one to four hours after spraying, from 1979 to 1982.

		Fenitrothion residues ug/g (dry weight)				Aminocarb residues ug/g (dry weight)			
	Number of samples	MEAN	MIN. - MAX.		MEDIAN	Number of samples	MEAN	MIN. - MAX.	MEDIAN
BALSAM FIR FOLIAGE									
1979	7	2.87	0.546	7.97	2.47	36	1.01	[1] N.D. 5.5	0.38
1980	35	12.01	0.006	111.0	5.38	6	9.82	0.522 37.3	4.53
1981	20	6.71	0.012	37.1	4.26	35	2.21	N.D. 26.2	0.98
1982	30	6.24	0.020	45.7	1.56	73	2.14	N.D. 20.0	1.09
1979-82	92	8.28	0.006	111.0	3.81	150	2.19	N.D. 37.3	0.94
		ug/ℓ				ug/ℓ			
LENTIC WATER									
1979	7	7.29	1.60	13.9	5.91	62	3.00	n.d. 25.0	0.70
1980	21	14.45	[2] n.d.	74.9	4.44	3	3.00	n.d. 7.95	1.04
1981	14	109.14	n.d.	583.0	6.05	32	3.46	n.d. 39.1	0.63
1982	21	71.08	0.09	1314.0	5.82	45	16.23	0.06 331.0	0.96
1979-82	63	53.57	n.d.	1314.0	5.82	142	7.29	n.d. 331.0	0.74
LOTIC WATER									
1979	7	20.52	0.02	127.0	1.18	34	1.22	n.d. 8.92	0.63
1980	36	11.07	0.01	124.0	3.74	6	2.40	0.06 6.95	1.13
1981	20	4.04	0.01	14.2	2.53	35	1.56	n.d. 18.4	0.79
1982	21	8.13	0.03	55.1	1.16	52	1.47	n.d. 6.46	1.11
1979-82	84	9.45	0.01	127.0	2.84	127	1.47	n.d. 18.4	0.86

[1] N.D. = not detected (<0.005 ug/g)

[2] n.d. = not detected (<0.010 ug/ℓ)

higher or lower than the median concentration for all four years. The only exception was the median aminocarb residue concentration in foliage sampled in 1980 which is 482% of the four-year median. This variation is probably due to the fact that only six samples were analyzed that year compared with a minimum of 35 samples for other years.

High residue levels found in lentic environments reaching 1,114 ug/l are probably due to the 585 diluent which renders the insecticide barely miscible with water and forms thin layers of oil on the surface of stagnent water. When displaced by the wind, these layers can be concentrated in certain places. In lotic environments, however, insecticides are diluted more easily by water movement and their recorded concentration levels are therefore generally lower. Only one study (Moody et al. 1978) mentions a high fenitrothion concentration (701 ug/l) in lotic water. However it should be noted that this sample was taken in a relatively still section of the stream (0.02 m/s). Another sample taken at the same time in the same stream, but where water flowed more rapidly (0.18 m/s), showed a concentration of only 1.32 ug/l.

Data mentioned in the literature available, which are provided in the introduction, are generally of the same magnitude as those described in this report. However, certain atypical concentrations detected in Quebec are much higher than the maximum levels mentioned in these studies. The size of the sampling network and the sampling method, which was designed to seek out maximum concentrations in the environment, may explain why very high residue levels were sporadically detected in Quebec.

The insecticide concentrations reported here were measured in foliage and water samples taken from one to four hours after spraying, when residue levels are likely to peak. In aquatic environments, fenitrothion residues disappear rapidly. Levels less than 1.0 ug/l were found two to eight days after spraying at 140 to 280 g AI/ha (Eidt and Sundaram 1975; Flannagan 1973; Lockhart et al. 1973; Mallet and Volpé 1982; Moody et al. 1978), and no trace of the insecticide could be found after 40 days (NRCC 1975). One hour after spraying fenitrothion at 210 g AI/ha in Quebec, an initial concentration of 543 ug/l was recorded in a pond. Within 3.3 hours, the concentration had diminished by 96%, and was only 0.346 ug/l after 16.5 days. Data available on aminocarb residue levels in natural bodies of water indicate that this insecticide dissipates quickly in aquatic environments (NRCC 1983).

On conifer foliage, fenitrothion has a half-life of two to four days; 70 to 95% of the residue dissipates in less than two weeks (Yule and Duffy 1972). A small proportion of the insecticide (< 10%) penetrates under the cuticle where it gathers (Yule 1974; Yule and Duffy 1972) and accumulates in subsequent years with continued spraying (McNeil et al. 1979; Yule 1974). Aminocarb also has a half-

life of only a few days in foliage (NRCC 1983) but minimal quantities can still be detected a year after treatment and sometimes even longer, depending on the minimum values detectable by the method of analysis (Mamarbachi 1980).

The median levels of residue detected in aquatic environments, which do not exceed 6 ug/l of fenitrothion and 1 ug/l of aminocarb, and their low persistence after spraying do not represent great hazards to aquatic fauna. Concentrations considered lethal to benthic invertebrate communities were estimated at 20 and 30 ug AI/l for operational mixtures of fenitrothion and aminocarb (Bio-Conseil Inc. 1981; Eidt and Weaver 1983). However, because residues are sometimes deposited unevenly, concentrations higher than the lethal threshold were sometimes recorded. These could affect aquatic invertebrates. In fact, data obtained from lotic water in Quebec show that 8.3% of the samples exceed 20 ug/l of fenitrothion and none exceed 30 ug/l of aminocarb. Some substantial increases in drifting aquatic invertebrates were even observed in streams after spraying (NRCC 1975). More serious impacts involve significant increase in the number of dead individuals and a significant drop in the density of benthic fauna (Symons 1977). Fenitrothion represents greater risks since concentrations measured during operational spraying are usually closer to known threshold values.

The safety margin is much greater for fish than aquatic invertebrates. The LC50/48 hours of a fenitrothion-oil spray mixture was estimated at 2,600 ug AI/l (Eco-Recherches Inc. 1982) for salmonids. A 48-hour exposure to 110 ug AI/l of this formula is required to reduce the salmonids' swimming performance by 30% (Eco-Recherches Inc. 1982). The levels at which lethal and sublethal effects on fish occur are much higher than those expected in either lentic or lotic waters after operational spraying. A review of toxicological data for freshwater organisms and environmental monitoring associated with spruce budworm control in Canada revealed no evidence of significant risk for aquatic environments if aminocarb (Matacil 180D™) is used as recommended (Holmes and Kingsbury 1980). Aminocarb residues expected in water after operational spraying in Quebec support this conclusion.

Insecticide residues detected in foliage are more likely to affect arthropods in trees but very few toxicological data are available for making comparisons. These organisms are usually killed by contact with the insecticide. Defoliator insects are most susceptible since they are intoxicated through contact and ingestion of contaminated foliage (Buckner 1974). Researchers (McNeil et al. 1979) have observed that fenitrothion residues of the order of 0.5 ug/g, which persist in conifer foliage, may cause unexpected disturbances in populations of Swaine Jack Pine Sawfly (Neodiprion Swainei Midd.), a non-target defoliator. However this species is one of the forest insect pests most susceptible to fenitrothion. The effect of fenitrothion and aminocarb regimes on predators found in fir stands has been studied for many years in New Brunswick. Conventional spray immediately kills many non-target arthropods but most taxa appear

resilient to such stress (Varty 1980). However, the hypothesis that the perennial use of these regimes may have an effect on population densities or diversities remains to be consolidated (Varty 1982).

By comparing our data concerning residual concentrations of fenitrothion and aminocarb, it appears that the contamination of water by the forest spraying of these insecticides presents few risks for fish and aquatic invertebrate populations. Immediate death of arthropods can be associated with the contamination of coniferous foliage but to date the impact on perennial population densities has not been established.

REFERENCES

Bio-Conseil Inc. (1981) Etude comparative des effets du matacil et du fénitrothion sur les populations benthiques en eaux courantes. Report prepared for the Min. de l'Energie et des Ressources du Québec, Québec, Canada

Buckner CH (1974) The biological side-effects of fenitrothion in forest ecosystems. Canadian Forestry Service, Report CC-X-67, Ottawa, Ontario, Canada

Eco-Recherches Inc. (1982) Effets toxiques d'une préparation opérationnelle de fénitrothion sur trois espèces de salmonidés. Report prepared for the Min. de l'Energie et des Ressources du Québec, Québec, Canada

Eidt DC, Sundaram KMS (1975) The insecticide fenitrothion in headwaters streams from large-scale forest spraying. Can Ent 107: 735-742

Eidt DC, Weaver CAA (1983) Threshold concentration of aminocarb that causes drift of stream insects. Can Ent 115: 715-716

Flannagan JF (1973) Field and laboratory studies of the effect of exposure to fenitrothion on freshwater aquatic invertebrates. Man Ent 7: 15-25

Holmes SB, Kingsbury PD (1980) The environmental impact of nonyl phenol and the matacil formulation, Part. 1: Aquatic ecosystems. Canadian Forestry Service, Sault Ste-Marie, Ontario, Canada

Holmes SB, Kingsbury PD, Mamarbachi G, Mathieu P (1984) Distribution of fenitrothion residues in Brook Trout (Salvelinus fontinalis) and Lake Trout (Salvelinus namaycush) tissues following aerial applications to Lac Ste-Anne, Québec, Bull Environ Contam Toxicol 33: 468-475

Lockhart WL, Metner DA, Grift N (1973) Biochemical and residue studies on Rainbow Trout (Salmo gairdneri) following field and laboratory exposures to fenitrothion. Man Ent 7: 26-35

Mallet VN, Cassista A (1984) Fenitrothion residue survey in relation to the 1981 spruce budworm spray program in New Brunswick, Canada. Bull Environ Contam Toxicol 32: 65-74

Mallet VN, Volpé G (1982) A chemical residue survey in relation to the 1980 spruce budworm spray program in New Brunswick (Canada). J Environ Sci Health 1317: 713-736

Mamarbachi G (1980) Recherche et dosage de l'aminocarbe dans les eaux naturelles et le feuillage du sapin baumier par chromatographie en phase gazeuse. Bull Environ Contam Toxicol 24: 415-422

McNeil JN, Greenhalg R, McLeod JM (1979) Persistence and accumulation of fenitrothion residues in Jack Pine foliage and their effect on the Swaine Jack Pine Sawfly, Neodiprion swainei. Environ Ent 8: 752-755

Moody RP, Greenhalgh R, Lockhart L, Weinberger P (1978) The fate of fenitrothion in an aquatic ecosystem. Bull Environ Contam Toxicol 19 (1): 8-14

National Research Council of Canada (1975) Fenitrothion: The effects of its use on environmental quality and its chemistry. Report No. 14104, Ottawa, Ontario, Canada

National Research Council of Canada (1983) Aminocarb: The effects of its use on the forest and the human environment. Report No. 18979, Ottawa, Ontario, Canada

Sundaram KMS, Szeto SY (1984) Persistence of aminocarb in balsam fir foliage, forest litter, and soil after aerial applications of three formulations. J Agric Food Chem Vol. 32 No 5: 1138-1141

Symons PEK (1977) Dispersal and toxicology of the insecticide fenitrothion; predicting hazards of forest spraying. Residue Rev 68: 1-36

Varty IW (1980) Effects on predaceous arthropods in trees. In: Varty IW (ed) Environmental surveillance in New Brunswick 1978-1979. University of New Brunswick, Fredericton, New Brunswick, p. 44

Varty IW (1982) Environmental impact of aerial spraying on predaceous arthropods in fir stands. In: Townsend, Washburn and Gillis Associates Ltd (ed) Environmental surveillance of spray operations for forest protection in New Brunswick, Fredericton, Canada, p. 70

Yule WN (1974) The persistence and fate of fenitrothion insecticide in a forest environment. II. Accumulation of residues in Balsam Fir foliage. Bull Environ Contam Toxicol 12 (2): 249-252

Yule WN, Duffy JR (1972) The persistence and fate of fenitrothion insecticide in a forest environment. Bull Environ Contam Toxicol 8 (10): 10-18

Yule WN, Varty IW (1975) The persistence and fate of fenitrothion insecticide in a forest environment. III. Deposit and residue studies with Black Spruce and Red Maple. Bull Environ Contam Toxicol 13 (6): 678-680

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